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# DUAL FLUIDIZED BED STEAM GASIFICATION OF COAL AND PYROLYZED COAL

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## ABSTRACT

The approaches for generating a tar free product gas by fluidized bed gasification are various. In this study the effect of volatile components in the feedstock on the gas quality was investigated. Therefore coal was pyrolyzed in a rotary kiln reactor before gasification in a dual fluidized bed steam gasifier. The results were compared in terms of gas quality and quantity for determination of the influence of the state of feedstock pyrolysis on the performance of steam gasification.

## INTRODUCTION

Gasification of coal represents a promising technology for upgrading a cheap feedstock to a high quality syngas for liquid and gaseous fuel production or for the synthesis of chemicals or other crude oil derived products like polymers. For the gasification of coal, most commercially available processes use air or a mixture of oxygen and steam as gasification agent. The production of oxygen is expensive and makes the process economically unattractive. If air is used for gasification the product gas is diluted with nitrogen and therefore not suitable for synthesis processes (1). When steam is used for gasification, the produced gas shows a high H<sub>2</sub> content and a high heating value, but the process becomes allothermal, so the heat for the gasification process has to be provided externally. The issue of heat supply for the gasification reactor is solved by using the innovative dual fluidized bed gasification technology (DFB). The DFB-technology separates the combustion reactor, which provides the energy for gasification, from the gasification reactor and pure steam is used as a gasification agent. Circulating bed material between these two reactors carries the heat from the combustion reactor to the gasification reactor. This gasification technology (2) has been developed at Vienna University of Technology, and has been successfully demonstrated, in Güssing and Oberwart, Austria, on the 8 and 10 MW<sub>th</sub> scale, respectively, since 2001 and 2008 (3). A high purity of the syngas, in particular concerning the tar content, is required for any downstream utilization of the gas especially for synthesis processes. If, for example, the product gas is used as a fuel for internal combustion engines, the tar content in the gas must not exceed 100 mg/Nm<sup>3</sup> (4). Moreover, the gas cleaning step has been identified to be one of the most expensive process steps. Therefore, the reduction of tars in the gasification reactor by primary measures is favored. As in the gasification reactor drying, pyrolysis and char gasification take place, the majority of the condensable products (tars) are formed by pyrolysis. This led to the idea to split the process into pyrolysis and char gasification. Thus, only the char was gasified in the fluidized bed reactor in order to limit the undesired tar species. To gain knowledge about the influence of pure char gasification on the gas quality and the performance, Polish hard coal, as the original feedstock, was pyrolyzed before gasification. The benchmark for the char gasification was the gasification of Polish hard coal, which was the origin of the char.

## MATERIALS AND METHODS

### The 3 MW pyrolysis pilot plant

For char generation, a rotary kiln pyrolysis pilot plant was used (5). This pyrolysis pilot plant has been designed for pyrolysis of agricultural residues but the reactor is able to handle a wide range of fuels due to its robust design and the long residence time of the feedstock in the kiln. The input fuel power for biomass is 3 MW, which corresponds to a fuel mass flow of around 650 kg/h for the standard feedstock. The pyrolysis reactor is a jacked rotary kiln reactor that is externally heated. The heating medium is hot gas that is produced in the afterburner by combustion of the pyrolysis gas. The scheme of the pyrolysis pilot plant can be found in (5).

### The 100 kW dual fluidized bed gasification pilot plant

For the experiments on pilot scale a dual fluidized bed gasification reactor, shown in Figure 1, is in operation at the Vienna University of Technology (VUT). A schematic drawing of the pilot rig is shown (2). This system separates gasification and combustion as two fluidized bed reactors connected together by loop seals are used. The fuel (coal, char) enters the gasification reactor, a bubbling bed fluidized with steam, where drying, pyrolysis and heterogeneous char gasification take place at bed temperatures of up to 900 °C. The remaining residual char leaves the gasification reactor at the bottom together with the bed material, which circulates between the two reactors. The combustion reactor is designed as a fast fluidized bed that is fluidized with air to maintain combustion of the residual char and additional fuel, if required. A more detailed description of the dual fluidized bed gasifier used at VUT can be found in (2).

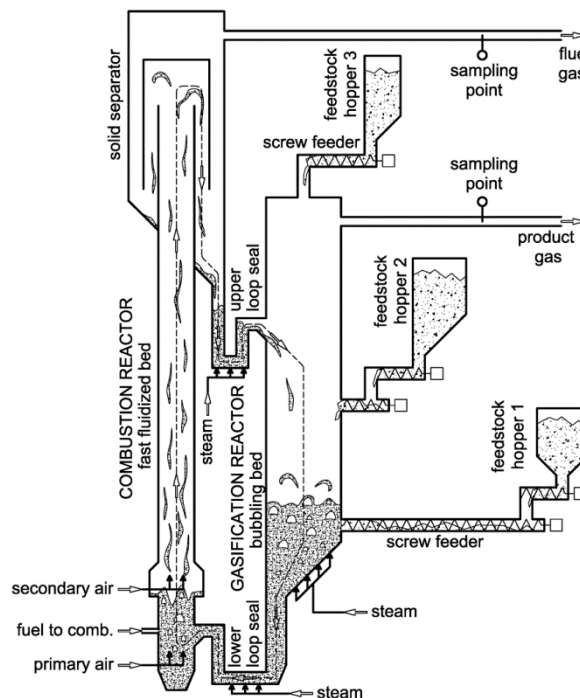


Figure 1: DFB gasification pilot plant

### Analysis

The composition of the product gas of the gasification process was measured after their exit from the reactor with the permanent gas components CH<sub>4</sub>, H<sub>2</sub>, CO, CO<sub>2</sub> and O<sub>2</sub> analyzed using a Rosemount NGA 2000, and N<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> via an online gas chromatograph (PerkinElmer Clarus 500). The tar measurement method is based on the tar protocol according to CEN/TS 15439 (6) focusing on tars originating from biomass gasification. The applied method here differs in the solvent used, as CEN/TS 15439 proposes isopropanol (IPA), but here toluene

was used. This allows a simultaneous determination of the water content in the product gas. This method was also used for measurement of tar in the pyrolysis gas which was produced during pyrolysis of the coal in the rotary kiln pyrolyzer. The condensable products formed during pyrolysis of the coal are called pyrolysis oil instead of tars due to their origin.

## RESULTS

### Coal pyrolysis

For production of char from Polish hard coal, the coal was pyrolyzed in the rotary kiln reactor. The pyrolysis temperature (outlet temperature of the pyrolysis gas) was chosen to 560°C. The key values for the process are summarized in Table 1.

**Table 1: Key values and results of the pyrolysis process**

| Value  | Unit                            | Rotary kiln pyrolysis |
|--|---------------------------------|-----------------------|
| Input mass flow rate coal                        | kg/h                            | 254.0                 |
| Output mass flow rate char                       | kg/h                            | 179.8                 |
| Pyrolysis gas <small>excl. pyrolysis oil</small> | Nm <sup>3</sup> /h              | 71.4                  |
| H <sub>2</sub> O content pyrolysis gas           | vol. %                          | 18.0                  |
| Content of entrained dust (inorganic)            | g/Nm <sup>3</sup> <sub>db</sub> | 2.11                  |
| Content of entrained char (organic)              | g/Nm <sup>3</sup> <sub>db</sub> | 9.3                   |
| Tar content gravimetric                          | g/Nm <sup>3</sup> <sub>db</sub> | 47.4                  |
| Tar content GC/MS                                | g/Nm <sup>3</sup> <sub>db</sub> | 18.5                  |

**Table 2: Proximate and ultimate analysis of the fuels**

|                 |       | Polish coal |         | Char from Polish coal |         |
|-----------------|-------|-------------|---------|-----------------------|---------|
|                 |       | dry basis   | as used | dry basis             | as used |
| Water content   |       | -           | 9.86    | -                     | 1.09    |
| Ash content     |       | 7.41        | 6.68    | 3.98                  | 3.94    |
| C               |       | 76.49       | 68.95   | 87.07                 | 86.12   |
| H               |       | 3.87        | 3.49    | 1.76                  | 1.74    |
| N               |       | 1.34        | 1.21    | 1.40                  | 1.38    |
| O               | wt. % | 10.29       | 9.26    | 5.45                  | 5.39    |
| S               |       | 0.46        | 0.41    | 0.25                  | 0.25    |
| Cl              |       | 0.15        | 0.14    | 0.09                  | 0.09    |
| Volatile matter |       | 34.66       | 31.24   | 7.48                  | 7.40    |
| Fixed carbon    |       | 65.34       | 58.90   | 92.52                 | 91.51   |
| LHV             | MJ/kg | 29.15       | 26.03   | 32.73                 | 32.35   |

By the pyrolysis process, mainly the volatile components and water were removed from the coal, so the fixed carbon content increased. As mentioned before the content of pyrolysis oil (tar) was also determined here with the same arrangement as it was used for the detection of tar in the product gas of the gasification process. Traditionally, much more condensable products are formed during pyrolysis compared to gasification at lower temperatures (higher temperature forces tar cracking reactions) and the missing gasifying agent. Nevertheless, the produced amount of pyrolysis oil was relatively low for the coal

pyrolysis here compared to other types of feedstock (5). This can be explained by the type of feedstock used in this study: the coal innately showed a low content of volatile components (Table 2), so the prospective yield of gaseous products was very low. The mechanism of slow pyrolysis was accentuated by this low gas yield. Nevertheless, increased gas residence time in the reactor offered the possibility of thermal destruction of the pyrolysis oil to non-condensable permanent gas components. Furthermore, the char can act as a catalyst in the reactor and produced pyrolysis gas can pass the char particles where adsorption and tar cracking of the condensable products (oil) will occur. The product of the pyrolysis process, the pyrolysis char, and its properties as well as the original feedstock, the Polish coal, are listed in Table 2.

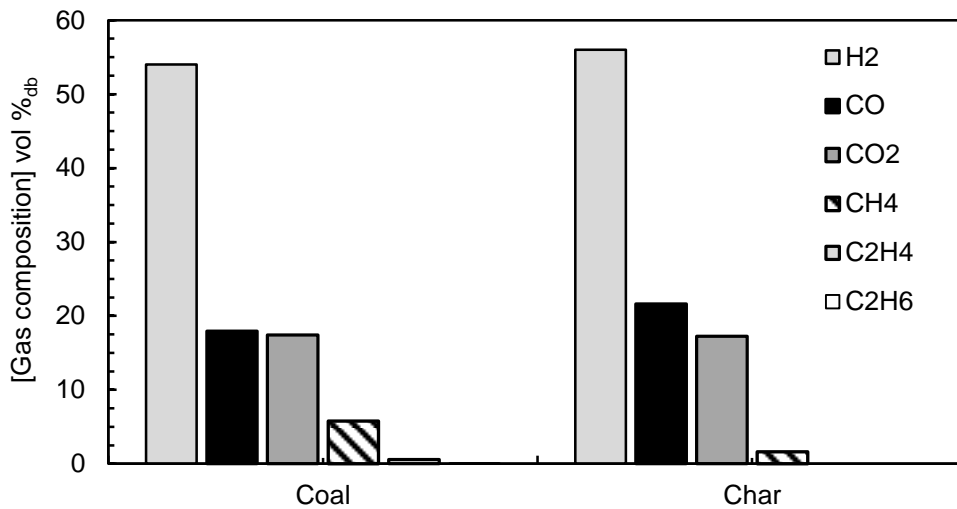
### Gasification of coal and pyrolyzed coal

The general parameters at which coal and char gasification was carried out in the DFB gasifier are summarized in Table 3. The two operating points differed only in terms of the feedstock, all other process parameters such as gasification temperature, bed material particle size and steam-to-carbon ratio ( $\varphi_{SC}$ ) have been kept constant. The gasification temperature was set to 870 °C while a fuel power (coal or char) of 78 kW was used. The bed material used in the reactor was calcined olivine. It was chosen as it shows a catalytic activity for tar reduction and is perceived as a non-toxic, natural catalyst. For each test in the pilot plant a new batch of olivine with an initial mass of 100 kg was used. The influence of the amount of steam as gasification agent in the gasification reactor is essential for system performance and product gas quality, so it is mandatory to maintain the same amount of steam for the gasification of solid carbon for both fuels. Therefore, the amount of steam introduced into the gasification reactor is referred to the introduced amount of fuel or to the introduced amount of carbon by the solid fuel. Those ratios are called the steam-to-fuel ratio ( $\varphi_{SF}$ ) and the steam-to-carbon ratio ( $\varphi_{SC}$ ) respectively.

**Table 3: Key data of the accomplished gasification tests**

| Value                                   | Unit                                      | Coal gasification | Char gasification |
|---|---|-------------------|-------------------|
| Gasification temperature (bubbling bed) | °C  | 870 ± 2           |                   |
| Temperature combustion reactor          | °C  | 926               | 912               |
| Fuel power                              | kW  | 78                |                   |
| Fuel mass flow (coal or char)           | kg/h                                      | 10.9              | 9.0               |
| Particle size bed material              | mm  | 0.4 - 0.6         |                   |
| Steam-to-carbon ratio, $\varphi_{SC}$   | kg <sub>H2O</sub> /kg <sub>carbon</sub>   | 1.9               | 2.0               |
| Steam-to-fuel ratio, $\varphi_{SF}$     | kg <sub>H2O</sub> /kg <sub>fuel,daf</sub> | 1.5               | 1.8               |

During the tests here, the steam-to-carbon ratio ( $\varphi_{SC}$ ) was kept nearly constant between 1.9 and 2.0 kg<sub>H2O</sub>/kg<sub>C</sub>. The change in the steam-to-fuel ratio ( $\varphi_{SF}$ ) towards higher values for char gasification was a consequence of the different feedstock mass flow rate to maintain the same input fuel power for all tests and the different carbon content of the fuels. However, based on previous experience it can be stated that the effects of these minor differences in the steam-to-fuel ratio on the process can be neglected.



**Figure 2: Measured permanent gas composition in the product gas**

Figure 2 shows the main product gas components at the outlet of the gasification reactor. There can be seen that for both fuels the H<sub>2</sub> content was relative high, but for char gasification this value increased up to 56.0 vol.%<sub>db</sub> compared to 54.0 vol.%<sub>db</sub> for coal gasification. This behavior can be explained as the char gasification reaction with H<sub>2</sub>O results in H<sub>2</sub> and CO and the increased carbon content of the char (Table 2) highlights this reaction. This fact was strengthened by the forced production of CO for char gasification: Also here the CO content increased from 18.0 vol.%<sub>db</sub> for coal gasification to 21.6 vol.%<sub>db</sub> for char gasification. The content of CO<sub>2</sub> was nearly not affected at all. Methane showed a significant decrease for char gasification. Here the CH<sub>4</sub> content decreased from 5.8 vol.%<sub>db</sub> down to 1.6 vol.%<sub>db</sub>. For the higher hydrocarbons C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> this effect was even more drastic as they vanished completely for char gasification. This showed that methane and the higher hydrocarbons are predominantly formed by devolatilization of the fuel particles while hydrogen and carbon monoxide is mainly a result of char gasification. This leads to the expectation of a significant decrease of tar in the product gas which will be shown in the following section. The water content in the product gas was between 46 and 49 vol.%, which was at a higher level than for standard operation with biomass due to the comparably high  $\varphi_{SC}$ . Due to these changes in the main product gas components, the lower heating value of the gas was also affected as the contribution of CH<sub>4</sub> and the higher hydrocarbons was missing for char gasification. Caused by this, the lower heating value of the product gas for char gasification was lower (9.61 MJ/Nm<sup>3</sup><sub>db</sub>) compared to that of coal gasification (10.97 MJ/Nm<sup>3</sup><sub>db</sub>). Inorganic (dust) as well as organic (char) matter entrained with the product gas were also measured. The results are shown in Figure 2. It can be seen that the dust content was nearly the same for both tests, but for char gasification the char content in the gas increased. This was explained by the higher content of fixed carbon of the feedstock and therefore, the higher content of char in the system. The tar content in the product gas was unquestioned one of the main objectives for these investigations. The gravimetric as well as the GC/MS detectable tars are plotted in Figure 2. The major finding here was that for char gasification nearly all of the tar components disappeared. For coal gasification 3.8 g/Nm<sup>3</sup><sub>db</sub> of gravimetric and

5.8 g/Nm<sup>3</sup><sub>db</sub> of GC/MS detectable tars were found, which are already lower values compared to biomass gasification (2).

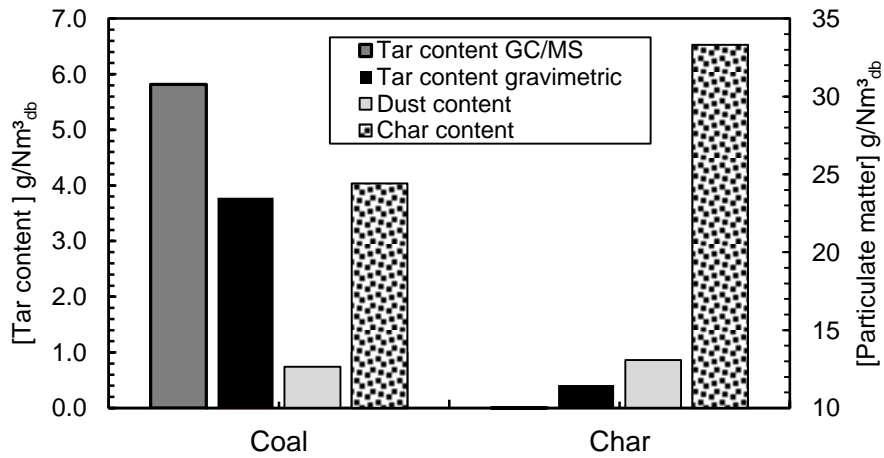


Figure 3: GC/MS and gravimetric tar content including particulate matter in the product gas

For char gasification the tar content regarding GC/MS tars was already in a range close to the detection limits of the method. Therefore, the sampling time was extended from 8 minutes (standard sampling) to 30 minutes for obtaining a sufficient amount of tar in the impinger bottles. For the GC/MS tar there can be assumed that no tar was present at all: the detected tar levels were between 1.0 and 3.0 mg/Nm<sup>3</sup><sub>db</sub> and only naphthalene was identified as tar component in the GC/MS system. A small amount of gravimetric tar was found with a mean value of 0.41 g/Nm<sup>3</sup><sub>db</sub>. However, a closer look on this value makes clear that this value was affected by the long sampling time and the high char and dust content in the product gas: a part of the high load of particulate matter was entrained through the filter cartridge to the toluene which was evaporated afterwards in a petri dish and by the weight of the tar that was collected (which was in this case diluted with some particles) was determined to calculate the tar content. This leads to the assumption that just some particles were weighted and the focus should be kept on the GC/MS detectable tars.

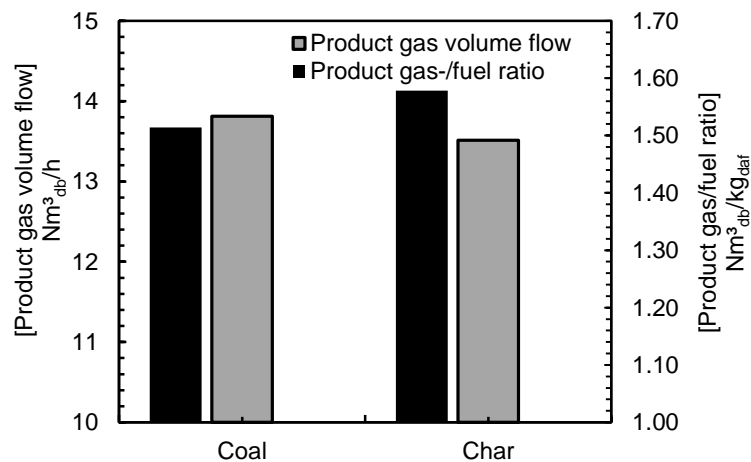


Figure 4: Absolute and specific gas production

The product gas quality was highly improved by the pre-pyrolysis process of the feedstock, but also the performance of the gasification process and the whole process chain should be considered. Figure 3 shows the quantitative production of product gas for both fuels. There can be seen that the total amount of product gas was higher for coal gasification but the specific product gas yield was higher for char gasification. However, for the comparison of coal and char in terms of product gas amount the values did not differ widely as the amount of volatile components was already quite low for the used coal. Beside the costs for the pyrolysis process the energy penalty during char production has to be kept in mind. While for the case where only coal was gasified in the system without thermal pretreatment, more than 53 % of the energy content bound in the coal was converted to product gas (Figure 4).

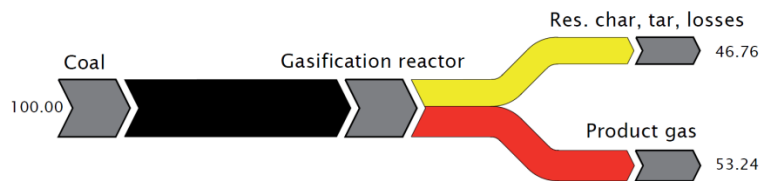


Figure 5: Split of the chemical feedstock energy via direct gasification (values in %)

In the case of char gasification there has to be the pyrolysis step considered (Figure 5) where already 12.2% of the initial energy content of the coal was removed and 87.5% of the energy of the coal was left in the char for gasification. Altogether the net production of gas by gasification is then only 39.1% of the original energy content in the feedstock. If also the pyrolysis gas with the high tar content can be used, then in total 51.3 % of the coal's energy can be used in form of gaseous products which nearly reached the values for direct coal gasification. Additionally, it has to be mentioned that the described tests refer to pilot plants with significantly higher heat losses than industrial scale plants. Thus, absolute numbers will be much higher then.

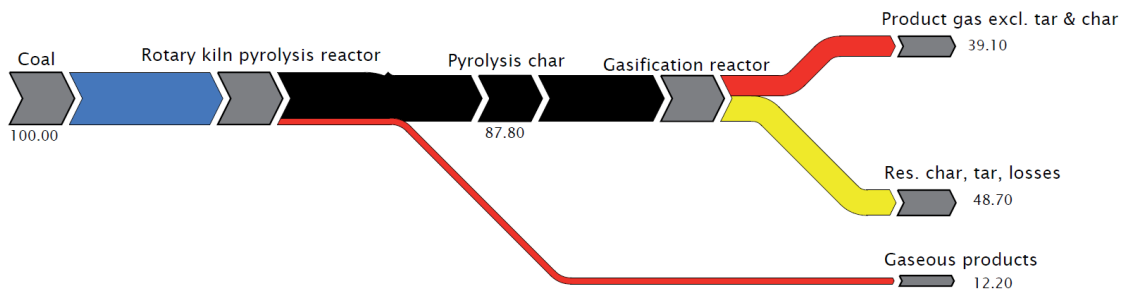


Figure 6: Split of the chemical feedstock energy via pyrolysis and char gasification (values in %)

## CONCLUSION

Pyrolysis as thermal pre-treatment process for coal gasification showed that the product gas quality was improved massively. The tar in the product gas vanished without any special in-bed catalyst and so a cost intensive tar removal system, can be avoided and the gas can be used directly in applications. Furthermore, for large scale plants the thermal energy can be recovered better by heat exchangers as it offers the possibility to cool down the gas to a lower level and fouling and slagging of the piping is avoided. The drawback with this method is the energy penalty by the additional upstream process step and that the carbon



conversion in the gasification reactor is lower for pure char gasification as more carbon is present in form of slowly reacting fixed carbon. However, a new dual fluidized bed gasifier design has been proposed (Z) which will enhance the contact with the hot bed material and therefore the carbon conversion will increase drastically. The tests presented here have shown one way to produce a tar free product gas by feedstock optimization without any system modification and it has been proven that the tar in the product gas made by steam gasification is formed mainly by volatile components of the feedstock.

## ACKNOWLEDGMENT

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## NOTATION

|                   |   |  |
|-------------------|---|--|
| $\varphi_{SF,wt}$ | steam-to-fuel ratio                               | $\text{kg}_{\text{H}_2\text{O}}/\text{kg}_{\text{fuel,daf}}$ |
| $\varphi_{SC,wt}$ | steam-to-carbon ratio                             | $\text{kg}_{\text{H}_2\text{O}}/\text{kg}_C$                 |
| CHP               | combined heat and power                           |  |
| daf               | dry and ash free basis                            |  |
| db                | dry basis   |  |
| DFB               | dual fluidized bed                                |  |
| GC/MS             | gas chromatography coupled with mass spectrometry |  |

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